Two-tag correlations and nonequilibrium fluctuation–response relation in ageing single-file diffusion

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Spatiotemporally correlated motions of interacting Brownian particles, confined in a narrow channel of infinite length, are studied in terms of statistical quantities involving two particles. A theoretical framework that allows analytical calculation of two-tag correlations is presented on the basis of the Dean-Kawasaki equation describing density fluctuations in colloidal systems. In the equilibrium case, the time-dependent Einstein relation holds between the two-tag displacement correlation and the response function corresponding to it, which is a manifestation of the fluctuation-dissipation theorem for the correlation of density fluctuations. While the standard procedure of closure approximation for nonlinear density fluctuations is known to be obstructed by inconsistency with the fluctuation-dissipation theorem, this difficulty is naturally avoided by switching from the standard Fourier representation of the density field to the label-based Fourier representation of the vacancy field. In the case of ageing dynamics started from equidistant lattice configuration, the time-dependent Einstein relation is violated, as the two-tag correlation depends on the waiting time for equilibration while the response function is not sensitive to it. Within linear approximation, however, there is a simple relation between the density (or vacancy) fluctuations and the corresponding response function, which is valid even if the system is out of equilibrium. This non-equilibrium fluctuation-response relation can be extended to the case of nonlinear fluctuations by means of closure approximation for the vacancy field.

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I. INTRODUCTION

Changes in systems tractable with statistical physics may occur endogenously as spontaneous fluctuations and exogenously in response to applied forces. In many cases, there is a relationship between these two types of changes [1, 2], which is known by the name of fluctuation–response relation (FRR) or, synonymously, fluctuation–dissipation relation. In particular, FRRs in systems near thermal equilibrium are well established and often referred to as the fluctuation–dissipation theorem (FDT) [1].

One of the simplest examples of FRR is the Einstein relation,

$$D = k_{\mathrm{B}}T/\mu, \tag{1.1}$$

between the diffusivity D and the drag coefficient μ of a free Brownian particle in a medium (typically water) with the temperature T. The diffusivity D measures how fast the mean square displacement (MSD) grows in time due to thermal fluctuations inside the system, while μ, or its inverse μ^{-1} (referred to as the mobility), represents the particle's response to external forces.

FRRs are useful in several ways. They help to understand the fluctuations of the system in terms of its response, and vice versa. Experimental verification of FRR serves as a test of basic assumptions underlying the theoretical model: a celebrated instance is found in Perrin's experiments [3] on the Einstein relation (1.1). Once FRR is established and written in the form of the Green–Kubo formula, it allows us to calculate transport coefficients, such as the viscosity, without applying shear in the calculation. In the (generalized) Langevin description of systems at thermal equilibrium with a fixed temperature, FDT prescribes the spectrum for the random force to play consistently the role of the heat bath with the temperature of the medium. FRR also concerns a certain type of theoretical approaches to correlations of fluctuations in nonlinear systems, known by the name of mode-coupling theory (MCT) [4–7] or direct-interaction approximation (DIA) [8, 9], as a kind of response function ("propagator") is involved in the procedure of the closure approximation.

To illustrate how FDT prescribes the random force term in the Langevin equation, let us consider a case of interacting Brownian particles. With the position vector of the i-th particle denoted with $\mathbf{r}_i = \mathbf{r}_i(t)$, the Langevin equation reads

$$m\ddot{\mathbf{r}}_i = -\mu\dot{\mathbf{r}}_i - \frac{\partial U}{\partial \mathbf{r}_i} + \mu\mathbf{f}_i(t) \quad (i = 1, 2, \ldots, N), \tag{1.2}$$

where $U = U(\{\mathbf{r}\}) = U(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N)$ denotes the interaction potential, and the drag term is assumed to be expressible with a constant scalar μ for the sake of simplicity. On the assumption that U does not affect the nature of the random force $\mathbf{f}_i(t)$, FDT requires [1]

$$\langle \mathbf{f}_i(t) \otimes \mathbf{f}_j(t') \rangle = \frac{2k_{\mathrm{B}}T}{\mu}\delta_{ij}\delta(t - t')\mathbb{1}, \tag{1.3}$$

where T is the temperature of the medium.

Generalization of FDT to nonequilibrium cases, with any appropriate modification, is one of the central problems of modern statistical physics. For example, for a class of nonequilibrium systems describable with the Langevin equation (1.2), the Harada–Sasa relation [10, 11] holds instead of the equilibrium FRR. In this case, the medium is in equilibrium so that the random force still satisfies (1.3), but if finite forcing included in U drives the

colloidal system out of equilibrium, the relation between the velocity autocorrelation of the particles and the system's response is modified. More generally, if a system exhibits FRR different from the equilibrium FDT, it manifests that the system is out of equilibrium. This violation of FDT has been observed in many nonequilibrium systems, such as glasses in ageing [12, 13], driven polymers [14], living cells [15, 16], and models of fluid turbulence [17–19]. Theoretical interpretation of an extra term in nonequilibrium FRR is also discussed as suggesting a role of novel statistical quantities, such as dynamical activity or "frenesy" [20, 21], corresponding to the role played by the entropy production in the near-equilibrium case.

In the context of transport in narrow channels, violation of the Einstein relation due to nonequilibrium initial configuration was recently discussed by Leibovich and Barkai [22]. They studied behavior of a tagged particle in a one-dimensional (1D) system of Brownian particles with hardcore interaction, which is a typical case of constrained dynamics known by the name of *single-file diffusion* (SFD) [22–29]. As the particles in such a system are hindered from free motion, they can diffuse only in some cooperative manner [30–32], with the MSD growing subdiffusively as

$$\left\langle R^2 \right\rangle \propto \sqrt{t} \tag{1.4}$$

for large t, where R is the displacement of the tagged particle in the time interval from 0 to t. Therefore, in SFD, the diffusivity in the usual sense vanishes; but it is still possible to define the time-dependent diffusivity and discuss its relationship with the time-dependent mobility [33], and thus the Einstein relation had been generalized to SFD and shown to be valid in the equilibrium case [33–35]. With this validity of the Einstein relation in the background, Leibovich and Barkai [22] compared two cases: ageing SFD started from the equidistant lattice configuration, and SFD with the initial condition already at equilibrium. The MSD was found to differ by the factor of $\sqrt{2}$, implying that the time-dependent diffusivity depends on the initial condition. Contrastively, the time-dependent mobility was found to be insensitive to the initial condition. In this way, the Einstein relation is violated in ageing SFD. An interesting point of this result is that the violation lasts forever, showing that it takes an infinitely long time to equilibrate the single-file system completely. This "everlasting effect" of the different initial conditions [22, 36] was recently shown to be produced also on higher-order moments of the displacement and on multi-time correlations, with a kind of Jepsen-like technique that makes a full use of the mapping from the SFD of point particles to non-interacting Brownian particles [37].

In this paper, we extend the work of Leibovich and Barkai [22] in several aspects. First of all, instead of MSD for only one tagged particle, we consider *two-tag* correlations to account for the cooperativity in SFD. We focus mostly on *displacement correlation* [32, 38, 39], denoting it with

$$\chi_{ij} = \langle R_i R_j \rangle \tag{1.5}$$

for the i-th and j-th particles. Correspondingly, the response function (i.e. the time-dependent mobility) is also treated as a two-body quantity g_{ij}. In relating χ_{ij} and g_{ij}, we also establish a connection with density fluctuations [39], beyond the linear approximation already known for a long time [26]. The approach to SFD based on density fluctuations (or, to be precise, fluctuations of elongation) has a wider applicability than the Jepsen-line approach, which means that we can generalize the result for point particles [22] to the case of particles with a finite diameter σ. While a linear analysis of density fluctuations suffices to reproduce the asymptotic behavior of MSD in (1.4), the nonlinear theory gives a subdominant term as a correction.

The paper is organized as follows. We start with background information about the time-dependent Einstein relation (subsection II.A), spatiotemporally correlated motions in SFD (subsection II.B), and description of the dynamics of interacting Brownian particles in terms of density fluctuations (subsection II.C). The equation for density fluctuations is called the Dean–Kawasaki equation. Subsequently, in section III, we specify the single-file system and define two-tag quantities such as χ_{ij}, g_{ij} and χ^{+}_{ij}, along with quantities in Fourier representations that bridge between these two-tag quantities and the Dean–Kawasaki equation. Main results are presented in sections IV and V: the former concerns the equilibrium case, while the ageing SFD is discussed in the latter. On the basis of density fluctuations in SFD, the time-dependent Einstein relation for MSD is generalized to the two-tag displacement correlation and the corresponding response function. The first main result is represented by Eq. (5.7), showing the same everlasting effect of the initial condition on the two-tag displacement correlation, as was found previously in one-tag cases [22, 36]. It is also discussed how to treat the effect of mode coupling due to the nonlinearity of the Dean–Kawasaki equation, using a closure approximation (MCT or DIA). While the standard approach is known to suffer from inconsistency with the FDT [5], this difficulty can be avoided with a suitable change of variables [38]. We apply this formalism to the non-equilibrium FRR with an extra ("frenesy") term [20, 40], showing that a closed relation between the correlation and the response is obtained without causing inconsistency. This is our second main result. Section VI is allotted for concluding remarks.

Some readers may find the background section too long, especially if they are already familiar with the time-dependent Einstein relation in SFD. In such a case, the readers would be advised to skip section II, except for figure 1 and the text around (2.20) which are necessary in the result sections. Readers who are more interested in the results than the methodology may also skim through section III, checking only important definitions such as (3.7), (3.13), (3.27) and (3.28), and then concentrate on sections IV and V.

II. BACKGROUND

A. Einstein relation

Before discussing SFD, let us begin with a brief review of the Einstein relation for free Brownian particles in the $n_{\rm d}$-dimensional space. By the word "free" we mean the case in which the interaction U is negligible in (1.2).

The diffusivity D represents *undriven* changes of $\mathbf{r}$ due to thermal fluctuations, through the definition

$$D = \lim_{t-s\to\infty} \frac{\langle [\mathbf{R}(t,s)]^2 \rangle}{2n_{\rm d}(t-s)}, \tag{2.1}$$

where $\mathbf{R}(t,s) = \mathbf{r}(t) - \mathbf{r}(s)$. Contrastively, application of a weak driving force ("probe force") to the same Brownian particle causes *driven* changes of its position, to be measured with the mobility μ^{-1}; if a constant probe force $\mathbf{F}^{\rm p}_{\infty}$ changes $\mathbf{r}(t)$ to $\mathbf{r}^{+}(t) = \mathbf{r}(t) + \Delta\mathbf{r}(t)$, the drift velocity is given by $(\mathrm{d}/\mathrm{d}t)\Delta\mathbf{r}(t) = (\mathrm{d}/\mathrm{d}t)\langle \mathbf{r}^{+}(t) \rangle$, which should be proportional to $\mathbf{F}^{\rm p}_{\infty}$ and therefore allows introducing μ such that

$$\frac{\mathrm{d}}{\mathrm{d}t} \left\langle \mathbf{r}^{+}(t) \right\rangle = \mu^{-1} \mathbf{F}^{\rm p}_{\infty} \tag{2.2}$$

in the steady state. The two constants defined in (2.1) and (2.2), namely D and μ, are connected by the Einstein relation (1.1). Note that (1.1) makes it possible to determine k_{B} experimentally and to evaluate thereby the Avogadro number, which played the historic role of demonstrating the reality of atoms [3].

More details on the motion of free Brownian particles may be given by solving the Langevin equation, i.e. (1.2) with $U = 0$. Let us utilize this example with $m > 0$ (though we will focus on the overdamped case in the remainder of this paper) to illustrate how the concept of mobility in (2.2) is generalized to the time-dependent response. For the sake of simplicity, we focus on the 1D case ($n_{\mathrm{d}} = 1$), rewriting (1.2) as

$$\left(m\frac{\mathrm{d}^2}{\mathrm{d}t^2} + \mu\frac{\mathrm{d}}{\mathrm{d}t}\right) X = \mu f(t), \tag{2.3}$$

with $X = X(t)$ denoting the position of the particle. With the initial values of X and $\dot{X} = v$ given at the time s, the solution for $t > s$ is

$$X(t) = X(s) + mv(s)\mathcal{G}(t-s) + \mu\int_s^t \mathrm{d}u\, \mathcal{G}(t-u)f(u) \tag{2.4}$$

where

$$\mathcal{G}(t') = \frac{1 - e^{-t'/\tau_{\mathrm{B}}}}{\mu} \qquad (t' > 0) \tag{2.5}$$

and $\tau_{\mathrm{B}} = m/\mu$. The function $\mathcal{G}$ represents the response of X to the forcing term on the right side of (2.3), which can be understood also as the response to the probe force in the following way. Suppose that an infinitesimal probe force $F^{\mathrm{p}} = F^{\mathrm{p}}(t)$, added to the right side of (2.3), changes the solution from X to X^+:

$$\left(m\frac{\mathrm{d}^2}{\mathrm{d}t^2} + \mu\frac{\mathrm{d}}{\mathrm{d}t}\right) X^+ = \mu f(t) + F^{\mathrm{p}}(t). \tag{2.6}$$

The change due to the probe force is expressible in terms of $\mathcal{G}$ given in (2.5), as

$$X^+(t) = X(t) + \int^t \mathrm{d}u\, \mathcal{G}(t-u)F^{\mathrm{p}}(u), \tag{2.7}$$

where the lower limit of the integral is understood as the time when the probe force is "switched on".

Now let us regard (2.7), instead of (2.5), as the definition of $\mathcal{G}$ that describes the response of X to F^{p}. By comparing (2.2) with (2.7) in the case of

$$F^{\mathrm{p}}(t) = \begin{cases} F^{\mathrm{p}}_\infty & (t > s) \\ 0 & (t < s) \end{cases} \tag{2.8}$$

and taking $(\mathrm{d}/\mathrm{d}t)\langle X(t)\rangle = 0$ into account, we find that the steady-state value of the response, $\mathcal{G}(+\infty)$, should be equal to the mobility μ^{-1}, as is readily verified by taking the limit of $t' \to \infty$ in (2.5). In other words, $\mathcal{G}$ can be regarded as a time-dependent generalization of the mobility.

To discuss the Einstein relation for $\mathcal{G}$, let us return to the solution (2.4) and calculate the MSD. Using the 1D version of (1.3) in regard to the noise statistics and averaging over the initial velocity $v(s)$ as well, we find

$$\left\langle R^2 \right\rangle = \frac{2k_{\mathrm{B}}T}{\mu}\left[t_\Delta - \tau_{\mathrm{B}}\left(1-e^{-t_\Delta/\tau_{\mathrm{B}}}\right)\right] + {\tau_{\mathrm{B}}}^2\left(\left\langle v^2\right\rangle_{\mathrm{init}} - \frac{k_{\mathrm{B}}T}{m}\right)\left(1-e^{-t_\Delta/\tau_{\mathrm{B}}}\right)^2, \tag{2.9}$$

where $R = R(t,s) = X(t) - X(s)$ and $t_\Delta = t - s > 0$. The mean value of $[v(s)]^2$, denoted with $\langle v^2\rangle_{\mathrm{init}}$, must be equal to $k_{\mathrm{B}}T/m$ at equilibrium, so that the last term in (2.9) vanishes. The derivative of the MSD with regard to t is then calculated as

$$\begin{aligned}\partial_t\left\langle [R(t,s)]^2\right\rangle &= \frac{2k_{\mathrm{B}}T}{\mu}\frac{\mathrm{d}}{\mathrm{d}t_\Delta}\left[t_\Delta - \tau_{\mathrm{B}}\left(1-e^{-t_\Delta/\tau_{\mathrm{B}}}\right)\right] \\ &= \frac{2k_{\mathrm{B}}T}{\mu}\left(1-e^{-t_\Delta/\tau_{\mathrm{B}}}\right);\end{aligned} \tag{2.10}$$

by comparing (2.10) with $\mathcal{G}$ in (2.5), we find

$$\partial_t\left\langle R^2\right\rangle = 2k_{\mathrm{B}}T\mathcal{G}(t-s), \tag{2.11}$$

which is an instance of the time-dependent version of the Einstein relation. Note that, if the response to the step force (2.8) is denoted with

$$\begin{aligned}\left\langle R^+(t,s)\right\rangle &= F_\infty^{\mathrm{p}}\int_s^t \mathcal{G}(t-u)\mathrm{d}u \\ &= F_\infty^{\mathrm{p}}\int_0^{t-s}\mathcal{G}(t')\mathrm{d}t',\end{aligned} \tag{2.12}$$

then (2.11) can be re-expressed as

$$\left\langle R^+(t,s)\right\rangle = \frac{\langle [R(t,s)]^2\rangle}{2k_{\mathrm{B}}T}F_\infty^{\mathrm{p}}. \tag{2.13}$$

It should also be noted that nonequilibrium initial condition, in which $\langle v^2\rangle_{\mathrm{init}}$ differs from $k_{\mathrm{B}}T/m$, results in violation of (2.13) on the timescale of τ_{B}.

Having reviewed the time-dependent Einstein relation, let us proceed to the case of interacting particles in SFD. The system is governed by the 1D version of (1.2) with short-ranged repulsive interaction potential, as will be specified later in section III. We consider the equation of motion in the overdamped limit ($m \to 0$), focusing on timescales greater than τ_{B}. Since SFD is subdiffusive, with the MSD behaving asymptotically as $\langle [R(t,s)]^2\rangle \propto \sqrt{t_\Delta}$ (where $t_\Delta = t-s$), the diffusivity vanishes according to the original definition in (2.1). Nevertheless, the time-dependent Einstein relation in the form of (2.13) is known to hold true for SFD at equilibrium [33–35]. For example, in the case of point particles with hardcore interaction, the MSD is given concretely in terms of the free-particle diffusivity $D = k_{\mathrm{B}}T/\mu$ and the mean density ρ_0 as

$$\left\langle R^2\right\rangle = \frac{2}{\rho_0}\sqrt{\frac{D\,t_\Delta}{\pi}}, \tag{2.14}$$

while the response of this system to the infinitesimal step force (2.8) is

$$\left\langle R^+\right\rangle = \frac{F_\infty^{\mathrm{p}}}{\rho_0 k_{\mathrm{B}}T}\sqrt{\frac{D\,t_\Delta}{\pi}}. \tag{2.15}$$

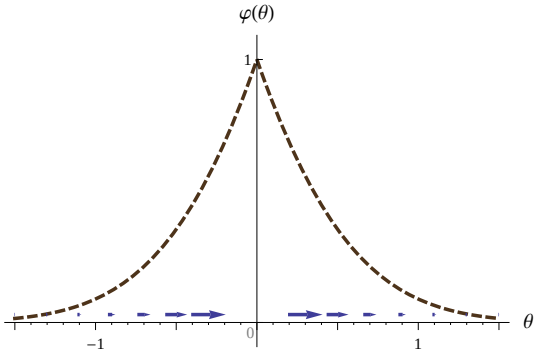

FIG. 1 The function $\varphi(\theta)$ in (2.20) describing the asymptotic behavior of the two-tag displacement correlation χ_{ij}.

Comparing (2.14) and (2.15), we notice readily that $\langle R^2 \rangle$ and $\langle R^+ \rangle$ satisfy the time-dependent Einstein relation in (2.13), because they are expressible as

$$\langle R^2 \rangle = K\sqrt{t_\Delta}, \qquad \langle R^+ \rangle = \frac{KF_\infty^{\mathrm{p}}}{2k_{\mathrm{B}}T}\sqrt{t_\Delta} \tag{2.16}$$

with the *same* constant K. This result holds true also for SFD of particles with finite diameter [35] and SFD on a lattice [33], with appropriate changes in K, as long as the system is at equilibrium. Contrastively, in the case of non-equilibrium SFD studied by Leibovich and Barkai [22], the Einstein relation (2.13) is violated.

The above-mentioned studies on (2.13) involve statistical quantities for a single tagged particle alone, such as the MSD. In SFD, however, we will find it more informative to understand the MSD as a limiting case of a two-tag correlation, taking the cooperativity into account. This is what motivates us to generalize the Einstein relation (2.13) to two-tag quantities.

B. Cooperativity in single-file diffusion

Let us consider SFD in a system at equilibrium, statistically homogeneous and steady, i.e. uniform both in space and in time. Although the subdiffusive behavior in (1.4) or (2.14) is probably the most remarkable feature of SFD, we emphasize the importance of another feature, referred to as *cooperativity*, which is more fundamental to the constrained dynamics in a narrow channel. It means that the particles in SFD are mutually caged and forbidden to move independently, so that their motions are possible only in some collective and cooperative manner. The subdiffusion in SFD is a consequence of the cooperativity in this sense and, in our opinion, should be understood as such. In [39], we have reviewed several approaches to SFD from this viewpoint, together with a historical example of experiments on K^+ transport across nerve cell membranes [30, 41], which impresses the importance of collective dynamics in narrow channels on us. It was this cooperativity that lead Hodgkin and Keynes [30] to the idea of a narrow channel for ions, forty years before crystallographic determination of ion channel structure.

Taking it for granted that the subdiffusion results from the cooperativity in SFD, one may develop different types of strategies. Some researchers prefer to eliminate the surrounding particles and reduce the collective single-file dynamics to a one-body problem described by a

generalized or fractional Langevin equation [35, 42–44]. In this description, the cooperativity is kept in the background, in the form of the memory kernel and the spatiotemporally correlated noise. Contrastively, here we present another type of strategy, targeting directly on the cooperativity.

Our main interest is in description and quantification of the collective dynamics with spatiotemporal correlations defined appropriately. Most basically, the collective dynamics in SFD are characterized by the dynamical correlation length $\lambda = \lambda(t)$, which describes the spatial length scale of the collective motion in the time interval from 0 to t (since the statistical steadiness is assumed, we can choose $s = 0$ without loss of generality, so that $t_\Delta = t$). In the case of finite systems, such as the ion channel with finite length L, the dynamical correlation length can span the entire system. In what follows, however, we will always assume that the system size L is so large that λ never reaches L. In such standard SFD with $L \to \infty$, the dynamical correlation length behaves asymptotically as

$$\lambda \propto \sqrt{t} \tag{2.17}$$

for large t, as will be explained below.

To prove (2.17), we notice that correlated motions in SFD are most conveniently quantified with the two-tag displacement correlation [32, 38]. On the assumption of time-translational and space-translational invariances, the two-tag displacement correlation χ_{ij}, given in (1.5), is a function of the "label distance" $\xi_{ij} = j - i$ (the particles are numbered consecutively) and the elapsed time t. This function $\chi_{ij} = \chi(\xi_{ij}, t)$ can be calculated by introducing a fluctuating field $h = h(\xi, t)$ that describes the positional fluctuation of the particles, with the "label variable" ξ being a continuum analogue of the particle numbering. As the crudest approximation, one may suppose that h behaves like a roughening surface subject to the Edwards–Wilkinson equation [45],

$$\partial_t h(\xi, t) = D' \partial_\xi^2 h(\xi, t) + f_h(\xi, t), \tag{2.18}$$

where f_h is a thermal noise such that[1]

$$\langle f_h(\xi, t) f_h(\xi', t') \rangle = 2D\delta(\xi - \xi')\delta(t - t').$$

By solving (2.18) in the Fourier representation, the two-tag displacement correlation is readily obtained [32, 38]:

$$\chi_{ij}(t) \propto \sqrt{t}\, \varphi\left(\frac{\xi_{ij}}{2\sqrt{D't}} \right), \tag{2.19}$$

with

$$\varphi(\theta) \stackrel{\text{def}}{=} e^{-\theta^2} - \sqrt{\pi}\, |\theta| \operatorname{erfc} |\theta| \,. \tag{2.20}$$

From (2.19) we can read the (nondimensionalized) dynamical correlation length $\lambda(t) = 2\sqrt{D't}$ that grows diffusively in proportion to $\sqrt{t}$. The function $\varphi(\,\cdot\,)$ is plotted in figure 1, where it is also shown as a 1D vector field, which may help intuitive understanding of the displacement correlation. Later, in section IV, we will refine the calculation of χ_{ij} on the basis of a nonlinear equation for density fluctuations, as opposed to the linear equation

[1] The coefficient D' in Eq. (2.18) may generally differ from D that determines the noise amplitude. See, for example, Eq. (4) in [35], where κ/ξ and $k_\mathrm{B}T/\xi$ corresponds to our D' and D.

(2.18). It will be shown that, in spite of the nonlinear effect, (2.19) remains valid for large t, so that (2.17) is proven.

The subdiffusive behavior of $\langle R^2 \rangle$ in (1.4) or (2.14) is a consequence of the growing correlation length. This is intuitively understood through Rallison's phenomenological argument [31], which could be reformulated in terms of the Einstein relation as follows: If n Brownian particles are strongly interacting and moving together, their center of mass behaves as a single Brownian particle with the mobility reduced by a factor of $1/n$ and therefore with the diffusion coefficient D/n, in the sense that

$$\frac{\mathrm{d}}{\mathrm{d}t} \left\langle R^2 \right\rangle = \frac{2D}{n}. \tag{2.21}$$

To apply (2.21) to the collective dynamics in SFD, Rallison [31] proposed to replace n in the denominator with $\mathcal{N}(\lambda) = 1 + \rho_0 \lambda$, which is the number of particles within the dynamical correlation length $\lambda = \lambda(t)$, so that the MSD is given by

$$\left\langle R^2 \right\rangle = \int_0 \frac{2D \, \mathrm{d}t}{\mathcal{N}(\lambda)}, \quad \lambda = \lambda_{\mathrm{Ral}}^{\mathrm{1D}}(t) = \sqrt{4\pi D t}. \tag{2.22}$$

Upon integration, (2.22) yields an expression that gives free diffusion for small t and reproduces (2.14) for large t. Thus the slowdown of diffusion is related to the growing number of particles in cooperative motion.

It should be noted that the correlation length $\lambda \sim \sqrt{Dt}$ differs significantly from the mean displacement of the particle, $\langle |R| \rangle \sim \sqrt{\langle R^2 \rangle}$. While it is obvious that displacement of a tagged particle requires cooperation of other particles at least within the covered distance $\langle |R| \rangle$ [46], it needs clarification why the actual number of cooperating particles, $\mathcal{N}(\lambda) \propto t^{1/2}$, is much greater than $\rho_0 \langle |R| \rangle \propto t^{1/4}$.

The mathematical origin of the diffusive t-dependence of λ can be traced back to the Edwards–Wilkinson equation (2.18). To understand the relevance of the Edwards–Wilkinson dynamics to SFD more physically, we suppose that the cooperative motion is mediated by some diffusing entity, whose dynamics is represented by the field h in (2.18). In the case of discrete SFD on a 1D lattice, the diffusing entity is identified as migrating vacancies [47]. The vacancy dynamics on the lattice have their counterpart in continuous SFD [38, 39, 48], in which the diffusing entity is the "free volume", i.e. fluctuation of the spatial interval between the particles. Within the linear approximation, the vacancy field (also known as the elongation field [49]) simply represents density fluctuations subject to a stochastic diffusion equation akin to (2.18), as will be explained in the next subsection.

Thus the cooperativity in SFD is grounded on the diffusive dynamics of density fluctuations. It is characterized by the dynamical correlation length $\lambda \sim \sqrt{Dt}$, and it results in the subdiffusive behavior of $\langle R^2 \rangle \propto t^{1/2}$, as the time-dependent diffusivity is reduced in inverse proportion to λ.

Before proceeding to the discussion on density fluctuation, some remarks on two-tag correlations in SFD may be in order here. The two-tag displacement correlation, $\chi_{ij} = \chi(\xi_{ij}, t)$, includes the MSD as its limiting case. Denoting the constant of proportionality in (2.19) again with K, we have

$$\chi_{ij} = K\sqrt{t}\, \varphi(\theta), \quad \theta = \frac{\xi_{ij}}{\lambda(t)}, \tag{2.23}$$

which implies $\langle R^2 \rangle = K\sqrt{t}$ because $\varphi(0) = 1$. It is interesting to note that χ_{ij} approaches $K\sqrt{t}$ even for $i \neq j$, if t is large enough to satisfy $|\xi_{ij}| \ll \lambda(t)$ so that the pair of tagged particles behaves as if a single particle. This result was recently generalized to arbitrary number of tags, by means of the vacancy dynamics on the lattice [50].

Besides the displacement correlation, some other forms of two-tag correlations are also known to be calculable. For the SFD of point particles, the probability distribution function for a rather general form of two-tag correlation can be calculated exactly [51]. The displacement correlation is obtained as one of its special cases, while another special case corresponds to the correlation of the inter-particle distance [35, 52]. Calculation of these correlations in SFD provides insight into their counterparts in higher dimensions, such as the displacement correlation tensor [53, 54] and the bond breaking correlation [55–57].

C. Fluctuating density of interacting Brownian particles

The explanation for basic features of SFD in the previous subsection was mostly based on the Edwards–Wilkinson equation (2.18). This is linked to the Langevin equation (1.2) through density fluctuations.

Let us consider a system of interacting Brownian particles subject to (1.2) in the n_{d}-dimensional space (later we will set $n_{\mathrm{d}} = 1$). Density fluctuations in this system can be described by a stochastic equation for the mesoscopic density field,

$$\rho = \rho(\mathbf{r}, t) = \sum_j \rho_j(\mathbf{r}, t), \tag{2.24}$$

where $\rho_j(\mathbf{r}, t) = \delta^{n_{\mathrm{d}}}(\mathbf{r} - \mathbf{r}_j(t))$. Since we focus on timescales greater than τ_{B}, the n_{d}-dimensional delta function $\delta^{n_{\mathrm{d}}}(\ \cdot\ )$ in the above expression should be regarded as a blunted one due to temporal coarse-graining.

The stochastic equation for ρ in this context is customarily referred to as the Dean–Kawasaki equation [7, 58–61]. Since ρ is conserved, the Dean–Kawasaki equation is most conveniently introduced as a set of two equations, consisting of the continuity equation

$$\partial_t \rho + \nabla \cdot \mathbf{Q} = 0 \tag{2.25}$$

and a stochastic equation for the flux,

$$\mathbf{Q} = -D\left(\nabla\rho + \frac{\rho}{k_{\mathrm{B}}T}\nabla U\right) + \sum_j \rho_j(\mathbf{r}, t)\mathbf{f}_j(t), \tag{2.26}$$

with the random forcing $\mathbf{f}_j(t)$ subject to (1.3). The term including

$$U = U[\rho](\mathbf{r}) = \int V_{\mathrm{eff}}(|\mathbf{r} - \tilde{\mathbf{r}}|)\rho(\tilde{\mathbf{r}})\mathrm{d}^{n_{\mathrm{d}}}\tilde{\mathbf{r}} \tag{2.27}$$

describes the interaction of the particles, with V_{eff} denoting the effective two-body potential, resulting from coarse-graining [59, 62] and expressed in terms of direct correlation function [59, 63, 64].

Elimination of $\mathbf{Q}$ from (2.25) and (2.26) yields a single equation which is "*almost* a closed equation [58]" for $\rho(\mathbf{r}, t)$. Instead of requiring too detailed information about every particle

in the random forcing term, $-\nabla\cdot\sum_j \rho_j(\mathbf{r},t)\mathbf{f}_j(t)$, one may introduce $f_\rho(\mathbf{r},t)$ whose statistics are prescribed as

$$\langle f_\rho(\mathbf{r},t)f_\rho(\mathbf{r}',t')\rangle = 2D\nabla\cdot\nabla'\rho(\mathbf{r},t)\delta(\mathbf{r}-\mathbf{r}')\delta(t-t'), \tag{2.28}$$

and replace the random forcing term with $f_\rho(\mathbf{r},t)$. The equation for $\rho(\mathbf{r},t)$ then reads

$$\partial_t\rho(\mathbf{r},t) = D\nabla\cdot\left(\nabla\rho + \frac{\rho}{k_\mathrm{B}T}\nabla U\right) + f_\rho(\mathbf{r},t). \tag{2.29}$$

Before discussing (2.29) as a nonlinear equation for ρ, let us review its linear approximation. Linearization of (2.29) around the mean density $\rho_0 = N/L^{n_\mathrm{d}}$ yields an equation for $\delta\rho(\mathbf{r},t) = \rho(\mathbf{r},t) - \rho_0$, which includes a convolution of V_eff and $\delta\rho$, analogous to (5.2) in [65]. The linearized equation is readily solved in the Fourier representation,

$$\hat\rho(\mathbf{k},t) = \int e^{\mathrm{i}\mathbf{k}\cdot\mathbf{r}}\delta\rho(\mathbf{r},t)\mathrm{d}^{n_\mathrm{d}}\mathbf{r} = \sum_j e^{\mathrm{i}\mathbf{k}\cdot\mathbf{r}_j(t)} \quad (\mathbf{k}\neq\mathbf{0}),$$

as it reads

$$\partial_t\hat\rho(\mathbf{k},t) = -D\mathbf{k}^2\left[1-\rho_0\hat c(k)\right]\hat\rho(\mathbf{k},t) + \hat f_\rho(\mathbf{k},t), \tag{2.30}$$

where $\hat c(k)$ is the Fourier transform of the direct correlation function $c(r)$, used to represent $V_\mathrm{eff}(r) = -k_\mathrm{B}Tc(r)$ and related to the static structure factor [66]

$$S(k) \overset{\mathrm{def}}{=} \frac{1}{N}\langle\hat\rho(\mathbf{k},t)\hat\rho(-\mathbf{k},t)\rangle = \frac{1}{1-\rho_0\hat c(k)}.$$

With the correlation of the Fourier modes defined as

$$\begin{aligned} F(k,t,s) &\overset{\mathrm{def}}{=} \frac{1}{N}\langle\hat\rho(\mathbf{k},t)\hat\rho(-\mathbf{k},s)\rangle \\ &= \frac{1}{N}\sum_{i,j}\left\langle e^{\mathrm{i}\mathbf{k}\cdot[\mathbf{r}_j(t)-\mathbf{r}_i(s)]}\right\rangle \quad (\mathbf{k}\neq\mathbf{0}), \end{aligned} \tag{2.31}$$

the linearized Dean–Kawasaki equation (2.30) yields

$$F(k,t,s) = S(k)e^{-D^\mathrm{c}k^2(t-s)}, \quad D^\mathrm{c} = D^\mathrm{c}(k) = \frac{D}{S(k)},$$

with $D^\mathrm{c}(k)$ referred to as the (short-time) collective diffusion coefficient [66]. Note that, if the factor $1-\rho\hat c(k)$ on the right side of (2.30) is approximated by a constant, this is simply a randomly forced diffusion equation (sometimes referred to as the diffusion-noise equation [42]), which has been proposed in many textbooks [67–69] independently of (2.29).

The Dean–Kawasaki equation (2.29) is often taken as a starting point for nonlinear theory of glassy dynamics [5, 7, 70–72]. The standard approach consists in an attempt to derive a nonlinear integrodifferential equation for F, referred to as MCT equation [6], using a systematic (field-theoretical) approximation. This attempt is obstructed, however, by the problem of inconsistency with the FDT. The difficulty occurs because nonlinearity arises not only from the interaction term $\rho\nabla U$ but also through f_ρ referred to as multiplicative noise [5, 58, 70–72], which means that ρ is present on the right side of (2.28). As it is difficult

to treat these two nonlinearities in a consistent manner, the standard expansion procedure leads to spurious violation of FDT and therefore failure in systematic derivation of the MCT equation from (2.29) [5].

Several ideas have been proposed to avoid this difficulty. While many researchers proposed to change the dependent variables [70–72], here we review the idea of changing the *independent* variable, which can be formulated in $n_{\rm d}$-dimensional systems [39] but seems to work most naturally in the 1D case [38, 48].

In the 1D case, the Dean–Kawasaki equation (2.29) governs the density field $\rho(x,t)$, with its flux $Q = Q(x,t)$ given by the 1D version of (2.26). The essential reason for adopting (x,t) as the independent variables is to perform coarse-graining; in other words, once the coarse-grained equation is obtained, there is no reason to stick to the variables (x,t). The idea is to change the independent variables back to (ξ,t), where ξ is the continuum analogue of the particle numbering, briefly mentioned in connection with (2.18) and referred to as the *label variable* [38, 39, 48]. The mapping from (x,t) to (ξ,t) is specified by the relation

$$\frac{\partial \xi}{\partial t} = -Q, \qquad \frac{\partial \xi}{\partial x} = \rho, \tag{2.32}$$

which implies

$$(\rho\partial_t + Q\partial_x)\,\xi(x,t) = 0$$

so that ξ is convected with the velocity $u = Q/\rho$, and thereby allows us to introduce ξ without numbering the particles explicitly. The inverse mapping from (ξ,t) to (x,t), plotted in the (ξ,x)-plane, is intuitively conceived as a roughening surface. The positional fluctuation, denoted by $h = h(\xi,t)$ in (2.18), is related with $x = x(\xi,t)$ as

$$h(\xi,t) = x(\xi,t) - \left\langle \frac{\partial x}{\partial \xi} \right\rangle \xi, \tag{2.33}$$

where $\langle \partial x/\partial\xi \rangle = \rho_0^{-1}$ is the mean slope of the "surface" $x = x(\xi,t)$. By differentiating (2.33) with regard to ξ and taking (2.32) into account, we find

$$\frac{\partial h}{\partial \xi} = \frac{\partial x}{\partial \xi} - \left\langle \frac{\partial x}{\partial \xi} \right\rangle = \frac{1}{\rho} - \frac{1}{\rho_0}. \tag{2.34}$$

Within linear approximation we have $\partial_\xi h \simeq -\rho_0^2 \delta\rho$, which establishes correspondence between the Edwards–Wilkinson equation (2.18) and the linearized Dean–Kawasaki equation. Besides, a systematic treatment of nonlinear effects is also possible [38], as we will see in the latter half of this article. The difficulty of FDT violation [5], rooted in the noise correlation (2.28) involving ρ and referred to as the multiplicative noise, is removed by the change of variables from (x,t) to (ξ,t), which expels ρ from the noise correlation as will be shown later in (3.23).

III. SETUP AND FORMULATION

A. Langevin equation for particles in a channel

Now let us specify the system studied here, though some important fragments are already given in the previous sections. We study a 1D system of Brownian particles, governed by the Langevin equation

$$\mu \dot{X}_i = F_i^{\rm int}(\{X\}) + \mu f_i(t), \tag{3.1}$$

which corresponds to the 1D version of (1.2) with $m \to 0$. The interaction between the particles is given by

$$F_i^{\rm int}(\{X\}) = -\frac{\partial}{\partial X_i} \sum_{j<k} V\left(|X_j - X_k|\right), \tag{3.2}$$

where $V(r)$ is a hardcore potential with a finite diameter σ, slightly mollified[2] for the convenience of numerical calculation. More specifically, we adopt

$$V(r) = \begin{cases} V_{\rm max}(1 - |r|/\sigma)^2 & (|r| \le \sigma) \\ 0 & (|r| > \sigma) \end{cases} \tag{3.3}$$

with $V_{\rm max}/k_{\rm B}T$ large enough to prevent overtaking. The random forcing is Gaussian with zero mean and

$$\langle f_i(t) f_j(t') \rangle = 2D\delta_{ij}\delta(t - t'), \tag{3.4}$$

where $D = k_{\rm B}T/\mu$.

The system consists of N particles, distributed homogeneously with the mean density $\rho_0 = N/L$, and the periodic boundary condition, $X_{i+N} = X_i + L$, is assumed. The system size is regarded as infinitely large ($L \to \infty$, $N \to \infty$). For later convenience, we define

$$\ell_0 \stackrel{\rm def}{=} \frac{L}{N} = \frac{1}{\rho_0} \tag{3.5}$$

to denote the mean distance $\langle X_{i+1} - X_i \rangle = \ell_0$.

The initial condition is assumed to be homogeneous, i.e. statistically uniform in space. Theoretically, it is specified through the static structure factor for the initial configuration, denoted with $S_{\rm init}$. In numerical calculations, we focus on the case of the equidistant configuration,

$$X_i|_{t=0} = i\ell_0 \quad (i = 0, 1, \ldots, N-1), \tag{3.6}$$

for which $S_{\rm init}(k) = 0$ all over the first Brillouin zone ($0 < |k| < \pi/\ell_0$).

B. Definition of statistical quantities

On the basis of the Langevin equation (3.1), here we define some statistical quantities involving two tagged particles. The average, denoted with $\langle \quad \rangle$, is taken over the random forcing in the Langevin equation and the initial condition, unless specified otherwise.

Note that the present system, governed by (3.1) without a driving force and started from a spatially uniform state, remains uniform in space and is invariant under the spatial reflection. This implies $({\rm d}/{\rm d}t)\langle X_i(t) \rangle = 0$; there is no drift on the average.

The most important role in the present work is played by the two-tag displacement correlation χ_{ij}. Although we have already given a rough definition in (1.5), here we restate

[2] This softening of V should not confused with the effective potential $V_{\rm eff}$ that appears in the Dean–Kawasaki equation through U in Eq. (2.27). A concrete form of $V_{\rm eff}$, corresponding to $V(r)$ with $V_{\rm max}/k_{\rm B}T \to +\infty$, will be given later in Eq. (3.21).

it somewhat more precisely. Denoting the displacement of the i-th particle for the time interval from s to t with $R_i(t,s) = X_i(t) - X_i(s)$, we define

$$\chi_{ij} = \chi_{ij}(t,s) \stackrel{\text{def}}{=} \langle R_i(t,s) R_j(t,s) \rangle \tag{3.7}$$

for a pair of particles labeled with i and j. The time order is assumed as $0 \le s < t$ and the particle numbering is consecutive. Although we could write $\chi_{i,j}$ instead of χ_{ij}, we make it a rule to omit a comma when possible.

To introduce a two-tag response function, which should appear in place of the mobility when the diffusivity in the Einstein relation is replaced by $\partial_t \chi_{ij}$, we add a small probe force (superscripted with p) to the particles governed by (3.1), so that the solution is changed from $\{X\}$ to $\{X^+\}$:

$$\mu \dot{X}_i^+ = F_i^{\text{int}}(\{X^+\}) + \mu f_i(t) + \mu f_i^{\text{p}}(t). \tag{3.8}$$

By expressing the change due to the probe force as

$$X_i^+(t) = X_i(t) + \int^t \mathrm{d}u \sum_j g_{ij}(t,u) f_j^{\text{p}}(u), \tag{3.9}$$

we define the (impulse) response function g_{ij}, with the lower limit of the integral understood in the same way as in (2.7). On averaging (3.9), denoting the lower limit with o and taking $\langle X_i(t) \rangle = \langle X_i(o) \rangle = \langle X_i^+(o) \rangle$ into account, we find

$$\left\langle X_i^+(t) - X_i^+(o) \right\rangle = \int_o^t \mathrm{d}u \sum_j \langle g_{ij}(t,u) \rangle \, f_j^{\text{p}}(u). \tag{3.10}$$

For the sake of simplicity, sometimes we omit $\langle \quad \rangle$ in expressions involving g_{ij} so that, for example, $g_{ij}(t,s)$ may actually mean $\langle g_{ij}(t,s) \rangle$. Note that, in the absence of the interaction through F_i^{int}, the response function g_{ij} would be reduced to the normalized hydrodynamic mobility matrix, which equals δ_{ij} in the present case.

In addition to g_{ij}, we introduce the step response by considering the probe force in the following form of a step function, applied to a specific particle, say the j-th one:

$$\mu f_i^{\text{p}}(t) = F^{\text{p}}(t) \delta_{ij} = \begin{cases} F_\infty^{\text{p}} & (t > s \text{ and } i = j) \\ 0 & (\text{otherwise}). \end{cases} \tag{3.11}$$

With $\langle R_i^{+(j)}(t,s) \rangle$ denoting the average displacement of the i-th particle caused by this step probe force, from (3.10) we find

$$\left\langle R_i^{+(j)}(t,s) \right\rangle = \frac{F_\infty^{\text{p}}}{\mu} \int_s^t g_{ij}(t,u) \, \mathrm{d}u. \tag{3.12}$$

It is then convenient to introduce the unit step response function,

$$\chi_{ij}^+(t,s) \stackrel{\text{def}}{=} \left\langle R_i^{+(j)}(t,s) \right\rangle / F_\infty^{\text{p}}, \tag{3.13}$$

which is related to the response function g_{ij} as

$$\chi_{ij}^+(t,s) = \mu^{-1} \int_s^t g_{ij}(t,u) \, \mathrm{d}u \tag{3.14}$$

and therefore also termed as the integrated response function.

With $\chi_{ij} = \langle R_i R_j \rangle$ and $\chi^+_{ij} = \langle R_i^{+(j)} \rangle / F^{\rm p}_\infty$ thus defined, we raise two questions about relationship between them. As was reviewed in the previous section, the time-dependent Einstein relation (2.13) holds between $\langle R^+ \rangle$ and $\langle R^2 \rangle$ concerning a single tagged particle in SFD. This relation can be rewritten as

$$2k_{\rm B}T\chi^+_{0,0}(t,s) = \chi_{0,0}(t,s), \tag{3.15}$$

where we have chosen $(i,j) = (0,0)$ to express the one-tag statistical quantities. The first question is whether (3.15) can be generalized to the cases of $i \neq j$ in SFD at equilibrium. The answer is affirmative, as will be shown in section IV on the basis of the Dean–Kawasaki equation. The second question concerns the effect of ageing on FRR, which will be discussed in section V.

Note that the space-translation invariance implies

$$\chi_{ij} = \chi_{i+m,j+m} = \chi_{0,j-i} \tag{3.16}$$

with m denoting here an arbitrary integer (not the mass). Analogous relations hold also for g_{ij} and χ^+_{ij}.

C. Label-based Fourier representation

In preparation for theoretical calculation of χ_{ij}, g_{ij} and χ^+_{ij} based on the Dean–Kawasaki equation (2.26), here we define the vacancy field $\psi = \psi(\xi,t)$ and introduce its Fourier modes, along with several statistical quantities related to them. Since overtaking is completely forbidden in the present case, with $V_{\max}/k_{\rm B}T \to +\infty$ in Eq. (3.3), the label variable ξ can be regarded simply as the continuous interpolation of the particle numbering. (Note, however, that the relation between ξ and the particle numbering can be somewhat more complicated in general [52].)

We define the vacancy field $\psi = \psi(\xi,t)$ as

$$\psi \stackrel{\rm def}{=} \frac{\rho_0}{\rho} - 1, \tag{3.17}$$

and regard it as continuum representation of

$$\psi_{i+1/2}(t) = \frac{X_{i+1}(t) - X_i(t) - \ell_0}{\ell_0}. \tag{3.18}$$

Note that ψ equals $\partial_\xi h$ in (2.34) up to a constant factor. With $u(\xi,t)$ denoting the velocity field, the vacancy field ψ is conserved in the sense that

$$\ell_0 \partial_t \psi(\xi,t) = \partial_\xi u(\xi,t), \tag{3.19}$$

which is a continuum representation of the time-derivative of (3.18):

$$\partial_t \psi_{i+1/2} = \frac{\dot{X}_{i+1} - \dot{X}_i}{\ell_0}. \tag{3.20}$$

With ψ thus introduced, we transform the Dean–Kawasaki equation into an equation for $\psi = \psi(\xi, t)$ [38, 48]. The procedure is outlined as follows: We substitute $u = Q/\rho$ into (3.19), with Q given by 1D version of (2.26), and adopt the effective potential

$$V_{\text{eff}}(r) \simeq k_{\text{B}}T \left[1 - \exp\left(-\frac{V(r)}{k_{\text{B}}T}\right)\right] = \begin{cases} k_{\text{B}}T & (|r| < \sigma) \\ 0 & (|r| > \sigma) \end{cases} \tag{3.21}$$

corresponding to the hardcore potential $V(r)$. Thereby we obtain [38, 48]

$$\ell_0 \partial_t \psi = -D\partial_\xi \left[\partial_\xi + 2\sinh\left(\frac{\rho_0 \sigma}{1+\psi}\partial_\xi\right)\right] \frac{\rho_0}{1+\psi} + f_{\text{L}}; \tag{3.22}$$

the thermal forcing term f_{L} is characterized by

$$\langle f_{\text{L}}(\xi, t) f_{\text{L}}(\xi', t')\rangle = 2D\partial_\xi \partial_{\xi'} \varpi(\xi)\delta(\xi - \xi')\delta(t - t'), \tag{3.23}$$

where $\varpi(\xi) = \sum_i \delta(\xi - i) \simeq 1$ (with the delta function blunted). Note an important difference between (2.28) and (3.23): while the noise correlation in the former depends on the unknown field ρ, this kind of dependence is expelled from (3.23), which implies that the difficulty due to the multiplicative noise [5] is basically removed here.

To make (3.22) more manageable, we switch to the Fourier representation (conjugate to ξ and marked with a hačeк), by defining

$$\check{\psi}(k, t) = \frac{1}{N}\int \mathrm{d}\xi\, e^{\mathrm{i}k\xi}\psi(\xi, t), \tag{3.24a}$$

$$\psi(\xi, t) = \sum_k \check{\psi}(k, t) e^{-\mathrm{i}k\xi}, \tag{3.24b}$$

where k is an integer multiple of $2\pi/N$. In this Fourier representation, (3.22) is rewritten as [38, 48]

$$\partial_t \check{\psi}(k, t) = -D^{\text{c}}_* k^2 \check{\psi}(k, t) + \sum_{p+q+k=0} \mathcal{V}^{pq}_k \check{\psi}(-p, t)\check{\psi}(-q, t) + \mathcal{O}(\check{\psi}^3) + \rho_0 \check{f}_{\text{L}}(k, t), \tag{3.25}$$

where

$$D^{\text{c}}_* = \frac{D_*}{S} = D_*\left(1 + \frac{2\sin\rho_0\sigma k}{k}\right), \qquad D_* = \rho_0^2 D$$

and

$$\mathcal{V}^{pq}_k = D_* k^2 W_{kpq} = D_* k^2 \left(1 + \frac{k}{pq}\sin\rho_0\sigma k + \frac{p}{kq}\sin\rho_0\sigma p + \frac{q}{kp}\sin\rho_0\sigma q\right). \tag{3.26}$$

Correlations of ψ in the Fourier representation are now introduced. Assuming $0 \le s < t$, we define

$$C(k, t, s) \overset{\text{def}}{=} \frac{N}{L^2}\left\langle \check{\psi}(k, t)\check{\psi}(-k, s)\right\rangle, \tag{3.27}$$

and we denote its single-time limit with

$$C^0(k, s) \overset{\text{def}}{=} \frac{N}{L^2}\left\langle \check{\psi}(k, s)\check{\psi}(-k, s)\right\rangle = \lim_{t\to s} C(k, t, s). \tag{3.28}$$

Note that the s-dependence of $C^0 = C^0(k,s)$ represents the ageing of the system in the present case.

The response function G, corresponding to C, is defined by adding a probe force term $\check{\Pi}(k,t)$ to (3.25) and expressing the change due to the probe force as

$$\check{\psi}^+(k,t) = \check{\psi}(k,t) + \int^t \mathrm{d}u \sum_{k'} G(k,t;k',u)\check{\Pi}(k',u), \tag{3.29}$$

with the notation in the pattern of (3.9) for X_i^+ and g_{ij}. To find correspondence between G and g_{ij}, we look for a link between $\check{\Pi}$ and f_i^{p}. This is found in (3.18) that connects ψ with X_i, implying a connection between (3.9) and (3.29) analogous to (3.20). By relating $\psi_{i+1/2}$ to $\check{\psi}(k,t)$ via the discrete Fourier transform

$$\check{\psi}(k,t) = \frac{1}{N}\sum_j e^{\mathrm{i}k(j+1/2)}\psi_{j+1/2}(t) \tag{3.30}$$

consistent with (3.24a), we find that $\check{\Pi}$ is given by the discrete Fourier transform of

$$\Pi_{i+1/2} \stackrel{\mathrm{def}}{=} \frac{f_{i+1}^{\mathrm{p}} - f_i^{\mathrm{p}}}{\ell_0}, \tag{3.31}$$

interpretable as a "tidal" probe force, i.e. a (discrete) gradient of the probe force field. The same discrete gradient of (3.9) in regard to i yields

$$\begin{aligned}\psi_{i+1/2}^+(t) &= \psi_{i+1/2}(t) + \int^t \mathrm{d}u \sum_j \left[g_{i+1,j}(t,u) - g_{i,j}(t,u)\right]\frac{f_j^{\mathrm{p}}(u)}{\ell_0} \\ &= \psi_{i+1/2}(t) + \int^t \mathrm{d}u \sum_j g_{ij}(t,u)\Pi_{j+1/2}(u),\end{aligned} \tag{3.32}$$

where we have utilized the space-translation invariance of g_{ij} in the form of (3.16). By comparing the discrete Fourier transform of (3.32) with the definition of G in (3.29), we find

$$\begin{aligned}G(k,t;k',s) &= \frac{1}{N}\sum_j\sum_m g_{jm}(t,s)e^{\mathrm{i}k(j+1/2)-\mathrm{i}k'(m+1/2)} \\ &= \delta_{k,k'}\sum_n g_{0,n}(t,s)e^{-\mathrm{i}kn}.\end{aligned} \tag{3.33}$$

Thus the diagonal components of $G(k,t;k',s)$, with $k = k'$, is linked to $g_{ij} = g_{0,j-i}$ by means of the discrete Fourier transform, and its off-diagonal components vanish. The inverse transform yields

$$g_{0,n}(t,s) = \frac{1}{N}\sum_k e^{\mathrm{i}kn}G(k,t,s), \tag{3.34}$$

where $G(k,t,s) = G(k,t;k,s)$ is a shorthand for the diagonal components.

D. Alexander–Pincus formula for displacement correlation

Now we complete this preparatory section with a formula to calculate the displacement correlation χ_{ij}.

It has been known for a long time [26] that the MSD in SFD can be expressed in terms of the correlation of density fluctuations, $F(k,t) \propto \langle \hat{\rho}(k,t)\hat{\rho}(-k,0)\rangle$, as

$$\left\langle R^2 \right\rangle \propto \int_{-\infty}^{+\infty} \frac{F(k,0) - F(k,t)}{k^2} \mathrm{d}k. \tag{3.35}$$

Upon substitution of $F(k,t) \simeq S(k)e^{-D^{\mathrm{c}}k^2 t}$, (3.35) readily yields $\langle R^2 \rangle \propto \sqrt{t}$. While (3.35) is valid only approximately, it is possible to improve on it by replacing $F(k,t)$ with the correlation of vacancy fluctuations, namely $C(k,t,s)$ in (3.27) [39]. We refer to (3.35), together with its improved versions, as the *Alexander–Pincus formula*.

Let us derive a variant of the Alexander–Pincus formula that allows calculation of χ_{ij} from C. By substituting (3.18) into (3.30), we find

$$\begin{aligned} \check{\psi}(k,t) &= \frac{1}{N} \sum_j e^{\mathrm{i}k(j+1/2)} \frac{X_{j+1}(t) - X_j(t) - \ell_0}{\ell_0} \\ &= \frac{e^{-\mathrm{i}k/2} - e^{\mathrm{i}k/2}}{N} \sum_j e^{\mathrm{i}kj} \frac{X_j(t) - j\ell_0}{\ell_0}, \end{aligned} \tag{3.36}$$

giving a linear relation between $\{\check{\psi}\}$ and $\{X\}$. This relation is readily inverted:

$$X_j(t) = X_{\mathrm{G}}(t) + j\ell_0 + \ell_0 \sum_{k\neq 0} \frac{e^{-\mathrm{i}kj}}{e^{-\mathrm{i}k/2} - e^{\mathrm{i}k/2}} \check{\psi}(k,t), \tag{3.37}$$

where X_{G} represents the center-of-mass motion. Since the system is assumed to be infinitely large, $X_j(t)$ in (3.37) is dominated by the long-wave modes, for which the denominator can be approximated with $-\mathrm{i}k$. The displacement is therefore

$$R_j(t,s) \simeq \ell_0 \sum_{k\neq 0} \frac{e^{-\mathrm{i}kj}}{-\mathrm{i}k} \left[\check{\psi}(k,t) - \check{\psi}(k,s)\right]. \tag{3.38}$$

Multiplying (3.38) by its duplicate, with (j,k) replaced by $(i,-k)$, taking the average and changing summation into integral, we arrive at the formula:

$$\chi_{ij}(t,s) = \frac{L^4}{\pi N^2} \int_{-\infty}^{+\infty} \left[\frac{C^0(k,t) + C^0(k,s)}{2} - C(k,t,s)\right] \frac{e^{-\mathrm{i}k\xi_{ij}}}{k^2} \mathrm{d}k, \tag{3.39}$$

where $\xi_{ij} = j - i$.

IV. SFD AT THERMAL EQUILIBRIUM

We are now ready to address the issue of two-tag FRR in SFD at thermal equilibrium. The question is whether the time-dependent Einstein relation between $\langle R^+ \rangle$ and $\langle R^2 \rangle$, in

the form of (2.13) or (3.15), can be generalized to a relation between the step response χ^{+}_{ij} and the displacement correlation χ_{ij} with $i \neq j$. Equivalently, we may discuss whether the impulse response g_{ij} is proportional to $\partial_t \chi_{ij}$.

We approach this problem by calculating C and G from the transformed Dean–Kawasaki equation (3.25) in the label-based Fourier representation. Since G is linked to g_{ij} by (3.34), and C to χ_{ij} by the Alexander–Pincus formula (3.39), we can thus discuss the relationship between g_{ij} and χ_{ij}.

A. Linear analysis of vacancy fluctuations

Let us begin with linear approximation to (3.25). The linearized equation reads

$$(\partial_t + D^{\mathrm{c}}_{*} k^2)\check{\psi}(k,t) = \rho_0 \check{f}_{\mathrm{L}}(k,t), \tag{4.1}$$

with the random force statistics given by the Fourier transform of (3.23),

$$\rho_0^2 \left\langle \check{f}_{\mathrm{L}}(k,t)\check{f}_{\mathrm{L}}(-k',t') \right\rangle = \frac{2D_*}{N} k^2 \delta_{kk'}\delta(t-t'). \tag{4.2}$$

An equation for $C(k,t,s)$ is obtained by multiplying (4.1) by $\check{\psi}(-k,s)$ and taking the average. Since $\left\langle \check{f}_{\mathrm{L}}(k,t)\check{\psi}(-k,s) \right\rangle$ vanishes due to the time order ($s < t$), we have

$$(\partial_t + D^{\mathrm{c}}_{*} k^2) C(k,t,s) = 0. \tag{4.3}$$

Similarly, for $C^0(k,s)$ we have

$$(\partial_s + 2D^{\mathrm{c}}_{*} k^2) C^0(k,s) = \frac{2D_* k^2}{L^2}, \tag{4.4}$$

where the expression on the right side originates from $\rho_0 \left\langle \check{f}_{\mathrm{L}}(k,s)\check{\psi}(-k,s) \right\rangle = D_* k^2/N$.

Since we assume now that the system is in a steady state at thermal equilibrium, $C^0(k,s)$ should be independent of s. As the steady solution to (4.4), we have

$$C^0 = \frac{S}{L^2} \quad \text{(in the steady state)}, \tag{4.5}$$

with the relation $D^{\mathrm{c}}_{*} = D_*/S$ taken into account. With (4.5) posed as the initial condition for C at $t = s$, the solution to (4.3) is

$$C(k,t,s) = \frac{S}{L^2} e^{-D^{\mathrm{c}}_{*} k^2 (t-s)}. \tag{4.6}$$

The two-tag displacement correlation χ_{ij} is calculated by substituting (4.6) into the Alexander–Pincus formula (3.39). Since the contribution from small k dominates, S and D^{c}_{*} can be safely replaced with their long-wave limiting values. For later convenience, we introduce

$$\begin{aligned} I_{\mathrm{EW}}(\xi,t') &\stackrel{\mathrm{def}}{=} \frac{1}{\pi}\int_{-\infty}^{+\infty} \frac{1 - e^{-D^{\mathrm{c}}_{*} k^2 t'}}{k^2} e^{-\mathrm{i}k\xi} \mathrm{d}k \\ &= 2\sqrt{\frac{D^{\mathrm{c}}_{*} t'}{\pi}}\, \varphi\left(\frac{\xi}{2\sqrt{D^{\mathrm{c}}_{*} t'}} \right), \end{aligned} \tag{4.7}$$

with the function $\varphi(\ \cdot\ )$ defined in (2.20). Thereby $\chi_{ij} = \chi_{ij}(t,s)$ is obtained from (4.6) as

$$\chi_{ij} = S\ell_0^2 I_{\mathrm{EW}}(\xi_{ij}, t_\Delta) = \frac{2S}{\rho_0}\sqrt{\frac{D^{\mathrm{c}} t_\Delta}{\pi}}\,\varphi\left(\frac{\xi_{ij}}{2\sqrt{D_*^{\mathrm{c}} t_\Delta}}\right), \tag{4.8}$$

where $\xi_{ij} = j - i$ and $t_\Delta = t - s$. Thus (2.19) is re-derived, with the coefficient reproducing quantitatively the MSD obtained by Kollmann [27].

The response function G satisfies essentially the same equation as C prescribing an exponential decay:

$$(\partial_t + D_*^{\mathrm{c}} k^2) G(k,t,s) = 0. \tag{4.9}$$

Solving (4.9) under the initial condition

$$G(k,t,s)\big|_{t=s} = 1, \tag{4.10}$$

we obtain

$$G(k,t,s) = e^{-D_*^{\mathrm{c}} k^2 (t-s)}. \tag{4.11}$$

This is then substituted into (3.34) and the sum is evaluated as an integral, which yields

$$g_{ij}(t,s) = \frac{1}{\sqrt{4\pi D_*^{\mathrm{c}} t_\Delta}} \exp\left(-\frac{\xi_{ij}^2}{4 D_*^{\mathrm{c}} t_\Delta}\right). \tag{4.12}$$

With g_{ij} and χ_{ij} thus obtained, it is easy to confirm

$$2D g_{ij}(t,s) = \partial_t \chi_{ij}(t,s). \tag{4.13}$$

Since g_{ij} is linked to χ_{ij}^+ by (3.14), the Einstein relation in (4.13) can be also written as

$$2k_{\mathrm{B}} T \chi_{ij}^+(t,s) = \chi_{ij}(t,s). \tag{4.14}$$

As a numerical test of these theoretical predictions, we calculated $\chi_{ij}(t,s)$ and $\chi_{ij}^+(t,s)$ for $(s,t) = (200, 300)\sigma^2/D$, from numerical solutions of (3.1) and (3.8). Since (4.8) and (4.14) predict

$$\frac{2k_{\mathrm{B}} T \chi_{ij}^+}{K\sqrt{t_\Delta}} = \frac{\chi_{ij}}{K\sqrt{t_\Delta}} = \varphi(\theta), \tag{4.15}$$

with

$$K = \frac{2S}{\rho_0}\sqrt{\frac{D^{\mathrm{c}}}{\pi}}, \qquad \theta = \frac{\xi_{ij}}{2\sqrt{D_*^{\mathrm{c}} t_\Delta}},$$

these equations can be tested by rescaling the numerical values of $\chi_{ij}^+(t,s)$ and $\chi_{ij}(t,s)$ in accordance with (4.15) and plotting them against θ. The plot is shown in figure 2, where the theoretical curve for $\varphi(\theta)$ is also included. The numerical data are consistent with (4.15) and thus support the Einstein relation between the two-tag quantities χ_{ij} and χ_{ij}^+.

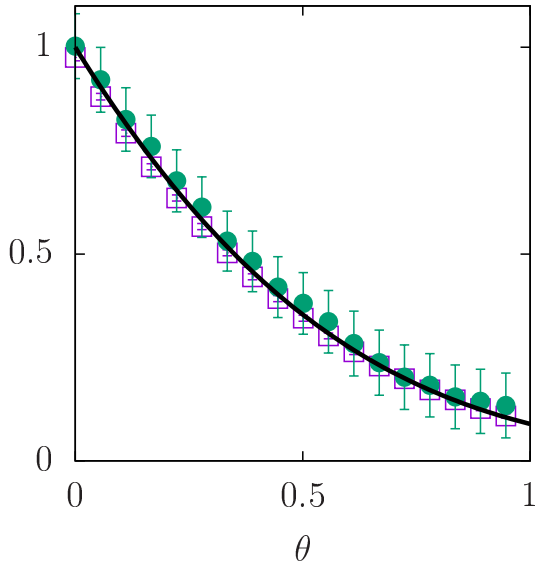

FIG. 2 Numerical verification of (4.15), based on the values of χ_{ij} and χ^{+}_{ij} calculated at $(s,t) = (200,300)\sigma^2/D$ from numerical solutions of (3.1) and (3.8), with $\rho_0 = N/L = 0.5\,\sigma^{-1}$. The symbols □ and • represent χ_{ij} and χ^{+}_{ij}, respectively, rescaled according to (4.15) and plotted against θ. The theoretical curve, $\varphi(\theta)$, is plotted with a solid line. Numerical parameters that should be infinite or infinitesimal are chosen as follows: $N = 1000$, $V_{\max} = 50\,k_{\mathrm{B}}T$, and $F^{\mathrm{p}}_{\infty} = 0.1\,k_{\mathrm{B}}T/\sigma$. The average was taken over 54400 runs in calculating χ^{+}_{ij}, while 1200 runs were sufficient in the case of χ_{ij} in which spatial averaging is also available. The static structure factor, needed for the rescaling, was evaluated numerically from equilibrium snapshots; the longwave limiting value was found to be $S \approx 0.31$ in the present case.

B. Nonlinear fluctuations and FDT

While some of the results in the previous subsection depend on the linear approximation, the Einstein relation in (4.13) or (4.14) must remain valid even if the nonlinear terms in (3.25) are taken into account. The FDT in systems at equilibrium is well established, and in the case of (3.25) it takes the form

$$\frac{D_* k^2}{L^2} G(k,t,s) = -\partial_t C(k,t,s), \tag{4.16}$$

as is seen heuristically by transforming (4.13) back into the relation between G and C. The concrete forms of G and C are modified by inclusion of the nonlinear terms, of course; in the next subsection, we will see how χ_{ij} is modified accordingly [38, 39].

The FDT in (4.16) can be derived from (3.25) by following an established procedure using the distribution function [2, 73]. To outline the derivation, we change the notation for a while to write (3.25) in the following form (sometimes called "model B" [74, 75]):

$$\partial_t \Psi_k(t) = -D_* k^2 \frac{\partial \mathcal{H}(\{\Psi\})}{\partial \Psi_k} + f_k(t) \tag{4.17}$$

with the variance of the thermal noise $f_k(t)$ being proportional to $2D_*k^2$. The thermodynamic potential $\mathcal{H}(\{\Psi\})$ has a minimum at $\{0\}$, toward which the system tends to relax.

The symmetry of W_{kpq} in (3.26) assures that (3.25) can be cast into the form of (4.17); we note that the original Dean–Kawasaki equation (2.29) is also expressible in terms of a free-energy functional of the density, but the relation between $\partial_t \rho(\mathbf{r}, t)$ and the free-energy functional is not so simple as (4.17) [58, 59].

The Langevin dynamics subject to (4.17) can be described by the Fokker–Planck equation (of Smolukowski type) [69, 76] that governs the distribution function, which we denote with $P = P(\{\Psi\}, t)$. In the steady state at thermal equilibrium, P is time-independent and equals

$$P^{\mathrm{eq}}(\{\Psi\}) \propto e^{-\mathcal{H}(\{\Psi\})}. \tag{4.18}$$

Following Falcioni *et al.* [73], we consider a change in the distribution function due to an impulsive probe force $\Pi_k = \epsilon\, \delta(t - s)$, applied to a specific mode k at the time s:

$$\begin{aligned} P^{+}(\{\Psi\}, t)\big|_{t=s+0} &= P^{\mathrm{eq}}(\ldots, \Psi_k - \epsilon, \ldots) \\ &= \left(1 + \epsilon \frac{\partial \mathcal{H}}{\partial \Psi_k}\right) P^{\mathrm{eq}} + \mathcal{O}(\epsilon^2), \end{aligned}$$

which implies

$$\begin{aligned} \frac{\left\langle \Psi_k^{+}(t) - \Psi_k(t) \right\rangle}{\epsilon} &= \left\langle \Psi(t) \left(\frac{\partial \mathcal{H}}{\partial \Psi_k}\bigg|_{\Psi(s)} \right) \right\rangle \\ &= \left\langle \Psi(s) \left(\frac{\partial \mathcal{H}}{\partial \Psi_k}\bigg|_{\Psi(t)} \right) \right\rangle \end{aligned} \tag{4.19}$$

with the time-reversal symmetry taken into account. The left side of (4.19) means G, while the expression on the left side turns out to be $-\partial_t C$ multiplied with a constant, with the aid of (4.17). Thus the FDT in (4.16) is derived from (4.17) at equilibrium, which was to be demonstrated.

C. Nonlinear theory: Lagrangian MCT

Keeping the FDT (4.16) in mind, let us present nonlinear calculation of C and χ_{ij} [38]. In this calculation we employ a closure approximation for C and G, known by the name of MCT or DIA. In contrast to derivation of MCT from the Dean–Kawasaki equation (2.29) for $\rho(\mathbf{r}, t)$ which suffers from FDT violation, the present derivation based on (3.25) is free from such a difficulty. We refer to the MCT for $\psi(\xi, t)$ as the Lagrangian MCT [38], using the terminology of fluid mechanics [9, 48, 77].

Let us start with writing an equation for $\partial_t C$ as a straightforward extension of (4.3). Instead of (4.1), we multiply (3.25) by $\check{\psi}(-k, s)$ and take the average, which yields an equation containing triple correlations:

$$\left(\partial_t + D_*^{\mathrm{c}} k^2\right) C(k, t, s) = \frac{N}{L^2} \sum_{p+q+k=0} \mathcal{V}_k^{pq} \left\langle \check{\psi}(-p, t) \check{\psi}(-q, t) \check{\psi}(-k, s) \right\rangle, \tag{4.20}$$

with the $\mathcal{O}(\check{\psi}^3)$ term in (3.25) discarded. With the aim of finding a closed set of equations that allows determination of C, the triple correlation on the right side is then expressed in

terms of memory integrals by prescription of DIA, which is based on the "sparseness" of $\mathcal{V}$ in Kraichnan's sense [8], as is briefly explained in Appendix B of [38]. This leads to a closed set of equations for C, C^0 and G:

$$\left(\partial_t + D_*^{\mathrm{c}} k^2\right) C(k,t,s) = \int_o^t \mathrm{d}u\, \tilde{M}^G(k,t,u) C(k,s,u) + \int_o^s \mathrm{d}u\, M^C(k,t,u) G(k,s,u) \tag{4.21}$$

$$\left(\partial_s + 2D_*^{\mathrm{c}} k^2\right) C^0(k,s) = 2\int_o^s \mathrm{d}u \left[\tilde{M}^G(k,s,u) C(k,s,u) + M^C(k,s,u) G(k,s,u)\right] + \frac{2D_* k^2}{L^2} \tag{4.22}$$

$$\left(\partial_t + D_*^{\mathrm{c}} k^2\right) G(k,t,s) = \int_s^t \mathrm{d}u\, \tilde{M}^G(k,t,u) G(k,u,s) \tag{4.23}$$

where

$$\tilde{M}^G(k,t,u) = M^G(k,t,u) - \Sigma(k,t)\delta_+(t-u) \tag{4.24}$$

$$M^G(k,t,u) = \frac{4L^2}{N} D_*^2 k^2 \sum W_{kpq}^2 C(p,t,u) q^2 G(q,t,u) \tag{4.25}$$

$$M^C(k,t,u) = \frac{2L^2}{N} D_*^2 k^4 \sum W_{kpq}^2 C(p,t,u) C(q,t,u). \tag{4.26}$$

We choose the initial time o to be either $o = 0$ or $o \to -\infty$; the latter is more convenient in the equilibrium case. For the most part we have followed the standard procedure of DIA [9, 38, 78, 79] (essentially equivalent to what is called the field-theoretical MCT), except for inclusion of

$$\Sigma(k,t) = \frac{L^2}{D_* k^2} M^C(k,t,t) \tag{4.27}$$

in (4.24), which mimics the effect of the higher-order correlation terms expected to cancel the singular short-time behavior of M^G; the symbol δ_+ represents a slightly shifted delta function such that

$$\int_0^\infty f(t)\delta_+(t)\mathrm{d}t = f(+0).$$

Now let us consider the equilibrium case, choosing $o \to -\infty$ and assuming time-translation invariance that allows us to write, for example, $C(k,t,s) = C(k,t_\Delta)$. In this case, it is demonstrated that (4.21), (4.22) and (4.23) are consistent with the FDT (4.16). By comparing the demonstration procedure in [38] with the corresponding attempt in the case of the original Dean–Kawasaki equation (2.29) [79], we find it essential that the coefficient of the nonlinear term has a symmetry as is seen in (3.26). The consistency with the FDT is recovered by the change of variable from $\rho(x,t)$ to $\psi(\xi,t)$, which has transformed the Dean–Kawasaki equation (2.29) and the multiplicative noise statistics (2.28) into the "model B" equation (3.25) with $\mathcal{V}_k^{pq}$ in (3.26) and the additive noise $\check{f}_{\mathrm{L}}$.

Once the consistency with the FDT is established, we can use (4.16) to eliminate G from (4.21), so that a closed equation for C is obtained:

$$\left(\partial_t + D_*^{\mathrm{c}} k^2\right) C(k,t-s) = -\int_s^t M(k,t-u)\partial_u C(k,u-s)\mathrm{d}u \tag{4.28}$$

where

$$M(k,u') = \frac{2L^4}{N} D_* k^2 \sum_{p+q+k=0} W_{pqk}^2 C(p,u') C(q,u'). \tag{4.29}$$

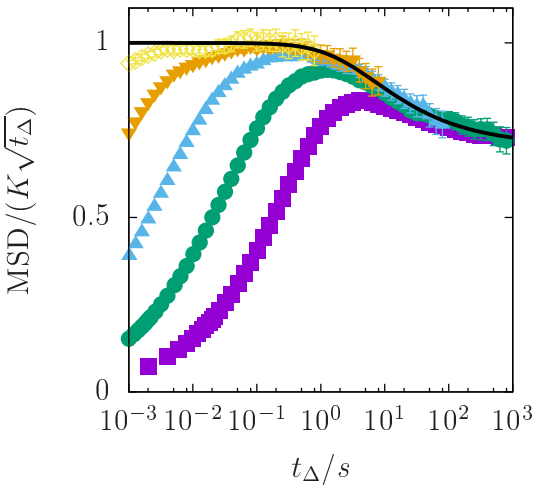

FIG. 3 Numerical values of the MSD in ageing SFD, plotted according to the scaling in (5.2). Given the equidistant initial condition (3.6), cases with five different values of waiting time are compared: $s = 1$ (■), 10, 10^2, 10^3 and 10^4 (◇) from bottom to top. The values of $\rho_0 = N/L$ and $V_{\max}$ are the same as in figure 2. The average was taken over 724 runs. The solid line shows $\mathcal{A}(t_\Delta/s)$ predicted by the linear theory [39, 80], a special case of (5.7) with $\xi_{ij} = 0$ and $S_{\text{init}} = 0$.

By solving (4.28) under the initial condition (4.5), we can obtain C and thereby calculate χ_{ij} via the Alexander–Pincus formula (3.39). An approximate solution for large t and vanishing σ is given in [38], from which χ_{ij} is calculated as

$$\chi_{ij} = 2\ell_0\sqrt{\frac{Dt_\Delta}{\pi}}\,\varphi(\theta) - \frac{\sqrt{2}}{3\pi}\ell_0^2\left(1-2\theta^2\right)e^{-\theta^2}, \tag{4.30}$$

where $\theta = \xi_{ij}/(2\sqrt{Dt_\Delta})$. The first term on the right side of (4.30) simply reproduces (4.8) with $S = 1$ (as $\rho_0\sigma$ is assumed to be vanishingly small), while the second term is a correction to it due to the entropic nonlinearity. A numerical test of (4.30) for $i = j$ is given in [39].

V. AGEING SFD

Now we are prepared to discuss the main issue of the present work: How are the two-tag FRRs in the previous section changed by ageing? Does the non-equilibrium initial condition with $S_{\text{init}} = 0$, whose effect on the MSD lasts forever according to Leibovich and Barkai [22], also affect the two-tag displacement correlation χ_{ij} in a similar way? The answer is affirmative, as is concretized in (5.7), which is the first main result of the present work.

We will start the discussion with comparing numerical plots of $\chi_{ij}(t,s)$ and $\chi^{+}_{ij}(t,s)$ in ageing SFD. Subsequently, the effect of ageing on these quantities will be clarified on the basis of the theoretical framework developed in the previous sections.

A. Numerical observations about effects of the waiting time

Leibovich and Barkai [22] reported an "everlasting effect of initial conditions" on SFD, comparing two extreme cases: SFD started from equidistant lattice configuration and SFD

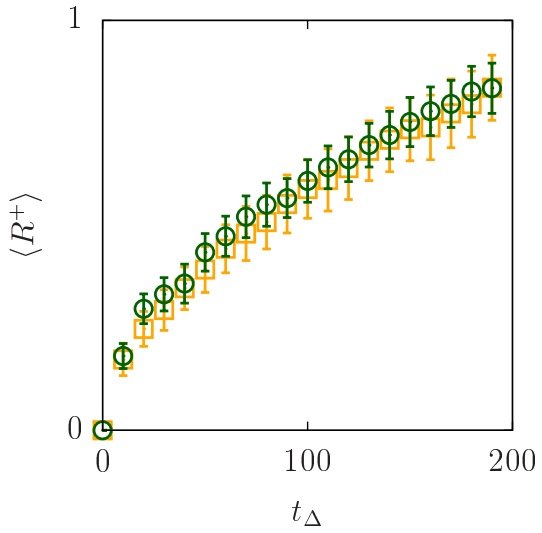

FIG. 4 The values of $\langle R^+ \rangle = \langle R^+(s + t_\Delta, s) \rangle$ plotted against t_Δ, with the space and time nondimensionalized by σ and σ^2/D. Two cases are shown: $s = 0$ ($\square$) and $s = 200$ ($\circ$). The average was taken over 25600 runs.

at equilibrium. These two cases can be treated in a unified manner by considering

$$\left\langle R^2 \right\rangle = \left\langle [R(t, s)]^2 \right\rangle = \left\langle [R(s + t_\Delta, s)]^2 \right\rangle \tag{5.1}$$

for different values of the "waiting time" s, under the equidistant initial condition (3.6). The two extreme cases correspond to $s = 0$ (equidistant lattice) and $s \to +\infty$ (equilibrated).

The MSD is calculated in this way from numerical solutions of (3.1) and shown in figure 3, where $\langle R^2 \rangle / (K\sqrt{t_\Delta})$ is plotted against t_Δ/s. As is predicted by the linear theory [39, 80], expected to be valid for $Dt_\Delta \gg \ell_0^2$, the MSD for different values of s is scaled in the form

$$\frac{\left\langle [R(s + t_\Delta, s)]^2 \right\rangle}{K\sqrt{t_\Delta}} = \mathcal{A}\left(\frac{t_\Delta}{s}\right) = \begin{cases} 1 & (t_\Delta \ll s) \\ \frac{1}{\sqrt{2}} & (t_\Delta \gg s). \end{cases} \tag{5.2}$$

The ratio of the values for the two extreme cases is consistent with the results by Leibovich and Barkai [22]. It is expressed in (5.2) that, however long the waiting time s may be, the effect of the non-equilibrium initial condition (3.6) reappears when t_Δ exceeds s; as a result, the MSD decreases by a factor of $1/\sqrt{2} \approx 0.71$.

Since the MSD thus depends on s, the Einstein relation (2.13) must be violated unless $\langle R^+ \rangle$ has the same s-dependence. Leibovich and Barkai [22] demonstrated, indeed, that $\langle R^+ \rangle$ is insensitive to s. This insensitivity is confirmed in figure 4: no significant difference is found between the numerical values of $\langle R^+ \rangle$ for $s = 0$ and that for $s = 200$. The Einstein relation (2.13) is therefore violated for $t_\Delta \gg s$.

To see how this violation of the Einstein relation is generalized to the two-tag case, in figure 5 we have plotted numerical values of $\chi_{ij}(s + t_\Delta, s)$ and $2\chi_{ij}^+(s + t_\Delta, s)$ against t_Δ, with nondimensionalization such that $k_\mathrm{B}T$, D and σ become unity. For $t_\Delta \ll s$, the plot in figure 5(a) obeys the two-tag Einstein relation (4.14), as the (nondimensionalized) values of χ_{ij} and $2\chi_{ij}^+$ coincide within the error bounds for all the three values of $j - i$ in the figure. As t_Δ increases toward the right end of figure 5(a), there seems to be a slight deviation. In the case of $s = 0$ shown in figure 5(b), the two-tag Einstein relation (4.14) is

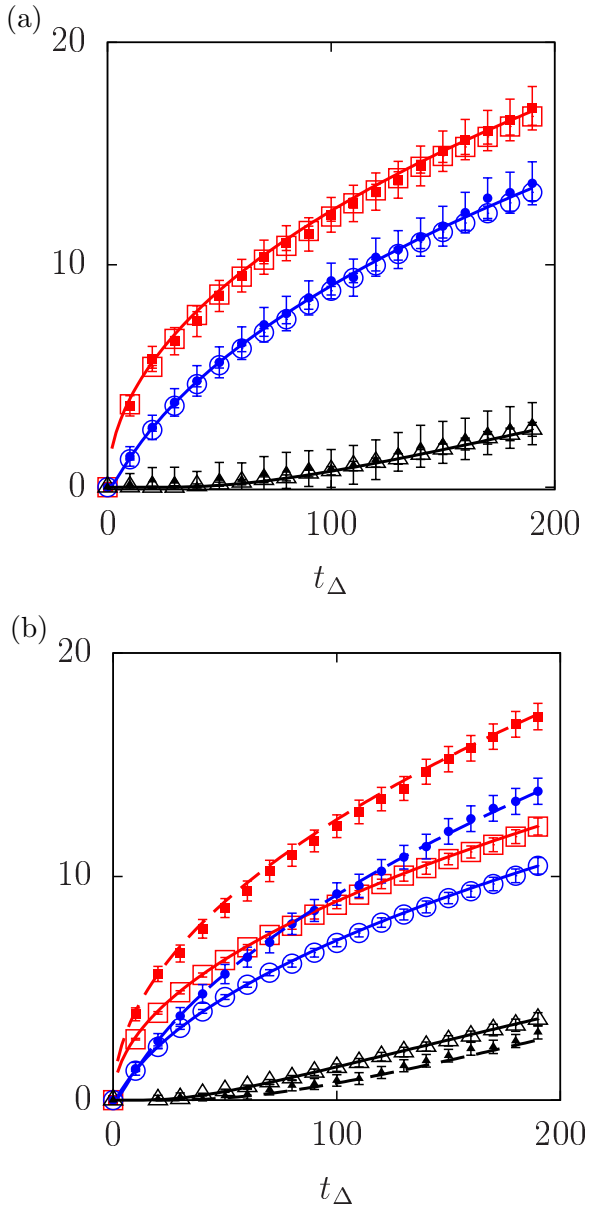

FIG. 5 Numerical tests of the two-tag Einstein relation between χ_{ij} and χ_{ij}^{+} and its violation. The open symbols □, ○ and △ represent $\chi_{ij}(s+t_\Delta, s)$ with $j-i=0$, 3 and 20, respectively; the filled symbols (■, ● and ▲) are used for the corresponding value of $2\chi_{ij}^{+}(s+t_\Delta, s)$. All the variables are nondimensionalized in units of $k_{\rm B}T$, D and σ. (a) The case of $s=200$, i.e. a waiting time longer than t_Δ. The theoretical curve for χ_{ij} in the equilibrium case, (4.8), is also shown. The average was taken over 400 runs for χ_{ij} and 76000 runs for χ_{ij}^{+}. (b) The case of $s=0$. The broken and solid lines represent the predictions of (4.8) and (5.7), respectively. The average for χ_{ij}^{+} was taken over 228000 runs.

evidently violated. Except for the short-time behavior in which χ_{ij} and $2\chi_{ij}^{+}$ are difficult to distinguish, the values of the displacement correlation χ_{ij} for $j=i$ and $j-i=3$ are smaller than the corresponding values of $2\chi_{ij}^{+}$. It is then interesting to note that, for $j-i=20$, the displacement correlation χ_{ij} is *greater* than $2\chi_{ij}^{+}$, implying that the ratio of χ_{ij} to χ_{ij}^{+} cannot be a function of (s,t) alone. This will be clarified as a part of analytical calculations in the

next subsection.

B. Linear analysis of ageing SFD

The s-dependence of χ_{ij} can be calculated analytically by applying the framework presented in section IV to the ageing case. Here we focus on linear analysis, with which concrete expressions for $\chi_{ij}(t,s)$ and $g_{ij}(t,s)$ are obtained.

Suppose that the system is initially in a state characterized by the static structure factor $S_{\text{init}}(k)$. In particular, the equidistant configuration (3.6) corresponds to $S_{\text{init}} = 0$. Subsequently, the system evolves according to (4.1) and relaxes toward the equilibrium, with $C^0(k,s)$ tending to its equilibrium value in (4.5). This is shown by solving (4.4) under the initial condition $C^0(k,0) = S_{\text{init}}/L^2$, which yields

$$C^0(k,s) = \frac{S}{L^2} + \frac{S_{\text{init}} - S}{L^2} e^{-2D_*^{\text{c}} k^2 s}. \tag{5.3}$$

Using (5.3) as the initial condition for C in (4.3) at $t = s$, we obtain

$$C(k,t,s) = \frac{S}{L^2} e^{-D_*^{\text{c}} k^2 (t-s)} + \frac{S_{\text{init}} - S}{L^2} e^{-D_*^{\text{c}} k^2 (t+s)}. \tag{5.4}$$

Note that (5.4) can be obtained also by solving (4.1) directly and using the noise amplitude in (4.2).

The response function G is found to be insensitive to ageing, within linear approximation, because (4.9) is independent of C and the initial condition for G, in (4.10), is also unchanged. This means that the solution in (4.11) remains valid.

Thus S_{init} has an effect on C^0 and C but not on G. Since G is linked to g by (3.34), we find that $g_{ij}(s + t_\Delta, s)$ is independent of s, being consistent with the observations of Leibovich and Barkai [22].

To calculate $\chi_{ij}(t,s)$, we substitute (5.4) into the Alexander–Pincus formula (3.39), which yields

$$\chi_{ij} = S\ell_0^2 I_1(\xi_{ij},t,s) + (S_{\text{init}} - S)\ell_0^2 I_2(\xi_{ij},t,s) \tag{5.5}$$

where

$$I_1(\xi_{ij},t,s) = I_{\text{EW}}(\xi_{ij}, t-s) \tag{5.6a}$$

$$I_2(\xi_{ij},t,s) = I_{\text{EW}}(\xi_{ij}, t+s) - \frac{1}{2}\left[I_{\text{EW}}(\xi_{ij},2t) + I_{\text{EW}}(\xi_{ij},2s)\right] \tag{5.6b}$$

with I_{EW} denoting the Edwards–Wilkinson integral (4.7). Therefore, using the function $\varphi(\ \cdot\ )$ defined in (2.20), we obtain

$$\begin{aligned}\chi_{ij}(t,s) = {}& \frac{2S}{\rho_0}\sqrt{\frac{D^{\text{c}}(t-s)}{\pi}}\,\varphi\!\left(\frac{\xi_{ij}}{2\sqrt{D_*^{\text{c}}(t-s)}}\right) \\ & + \frac{S_{\text{init}} - S}{\rho_0}\left[2\sqrt{\frac{D^{\text{c}}(t+s)}{\pi}}\,\varphi\!\left(\frac{\xi_{ij}}{2\sqrt{D_*^{\text{c}}(t+s)}}\right)\right. \\ & \left. - \sqrt{\frac{2D^{\text{c}}t}{\pi}}\,\varphi\!\left(\frac{\xi_{ij}}{2\sqrt{2D_*^{\text{c}}t}}\right) - \sqrt{\frac{2D^{\text{c}}s}{\pi}}\,\varphi\!\left(\frac{\xi_{ij}}{2\sqrt{2D_*^{\text{c}}s}}\right)\right].\end{aligned} \tag{5.7}$$

The first term on the right side of (5.7) is a function of t_Δ reproducing the equilibrium result in (4.8), while the other term, proportional to $S - S_{\rm init}$ and irreducible to a function of t_Δ alone, expresses the ageing effect. The Einstein relation (4.13) is violated because there is no change in g_{ij} corresponding to the ageing term in (5.7)

For MSD ($\xi_{ij} = 0$), (5.7) can be reexpressed in the scaling form of (5.2) [80],

$$\frac{\langle [R(s+t_\Delta, s)]^2 \rangle}{K\sqrt{t_\Delta}} = 1 + \frac{S_{\rm init} - S}{S} \left(\sqrt{1 + \frac{2s}{t_\Delta}} - \sqrt{\frac{1 + s/t_\Delta}{2}} - \sqrt{\frac{s}{2t_\Delta}} \right). \tag{5.8}$$

It is easy to check consistency with Leibovich and Barkai [22] by calculating the two limiting values for $s \ll t_\Delta$ and $s \gg t_\Delta$. The former limit ($s \ll t_\Delta$) is readily obtained by setting $s = 0$ in (5.8), which yields, after some rearrangements,

$$\frac{\langle [R(t_\Delta, 0)]^2 \rangle}{K\sqrt{t_\Delta}} = \frac{S + (\sqrt{2} - 1) S_{\rm init}}{\sqrt{2}\, S}. \tag{5.9}$$

This interpolates the case of the equidistant initial condition ($S_{\rm init} = 0$) and the equilibrium case ($S_{\rm init} = S$), reproducing the factor of $1/\sqrt{2}$. Our result in (5.9) is also consistent with (13) in [36], if $T_{\rm chain}/T$ is interpreted as $S_{\rm init}/S$. Note, however, that the chain temperature $T_{\rm chain}$ cannot be used to characterize distribution of hardcore particles, for which $S_{\rm init}$ seems to be more appropriate. In the opposite limit of $s \gg t_\Delta$, the expression on right side of (5.8) tends to unity (i.e. the same value as in the equilibrium case), as it ought to be.

Although (5.7) for $\xi_{ij} \neq 0$ seems rather complicated, a considerable simplification is possible in the limit of $s \to 0$. We have

$$\chi_{ij}(t, 0) = \frac{S}{\rho_0} \sqrt{\frac{2D^{\rm c} t}{\pi}}\, \varphi\left(\frac{\xi_{ij}}{2\sqrt{2D^{\rm c}_* t}} \right) \tag{5.10}$$

in this limit (we have set $S_{\rm init} = 0$ for further simplification). Comparing (5.10) with the equilibrium result in (4.8), we find two differences: the amplitude is smaller by a factor of $1/\sqrt{2}$, while the correlation length is *longer*. It means that $\chi_{ij}(t_\Delta, 0)$ for large ξ_{ij} can be *greater* in comparison to the corresponding equilibrium value, $\lim_{s\to+\infty} \chi_{ij}(s + t_\Delta, s)$, in contrast to the behavior near $\xi_{ij} = 0$. This is consistent with the numerical result for $s = 0$ shown in figure 5, with the insensitivity of $\chi^+_{ij}(s + t_\Delta, s)$ to s taken into account. Thus the ratio of χ_{ij} to χ^+_{ij}, which would equal $2k_{\rm B}T$ in equilibrium, is found to depend not only on (t, s) but also on $j - i$ in the present case of ageing SFD.

C. Non-equilibrium FRR

To close the discussion, let us derive a non-equilibrium FRR between G and C. Since (4.13) is violated, (4.14) also ceases to hold, and it seems difficult to find a simple relation between $\chi_{ij}(t, s)$ and $\chi^+_{ij}(t, s)$ in the non-equilibrium case. However, in regard to G and C in (4.11) and (5.4), there is a simple relation:

$$G(k, t, s) = \frac{L^2}{2D_* k^2} (\partial_s - \partial_t) C(k, t, s). \tag{5.11}$$

This is a linearized form of the non-equilibrium FRR between G and C explained below. The nonequilibrium FRR, including nonlinear fluctuations, is shown to be tractable with DIA; this methodological insight is our second main result.

Derivation of the nonlinear FRR for (3.25) may be discussed as an instance of generic methods for FRR in ageing systems governed by a nonlinear equation of Langevin type [11, 40]. The existence of a steady distribution function, which played a pivotal role in the derivation of the equilibrium FDT (4.16) in section IV, cannot be assumed in such systems. Instead, we can rely on another assumption that the "water" is always at equilibrium so that the fluctuating force is Gaussian.

To outline the idea for non-equilibrium FRR in such systems [40], here we write (3.25) or (4.17) even more schematically as

$$\partial_t \psi_t = \mathcal{F}(\psi_t) + f_t, \qquad \langle f_t f_{t'} \rangle = 2\mathcal{T}\delta(t-t'), \tag{5.12}$$

where the k-dependence is omitted and the time arguments of ψ and f are subscripted. Defining $C(t,s) = \langle \psi_t \psi_s \rangle$ with the time order $s < t$, we have

$$\partial_t C(t,s) = \langle \mathcal{F}(\psi_t)\psi_s \rangle + 0 \tag{5.13}$$

$$\partial_s C(t,s) = \langle \psi_t \mathcal{F}(\psi_s) \rangle + \langle \psi_t f_s \rangle \,. \tag{5.14}$$

Due to the Gaussian property of f, the last term in (5.14) is known to give the response function $G(t,s)$ [2, 81]:

$$\langle \psi_t f_s \rangle = 2\mathcal{T} G(t,s), \tag{5.15}$$

which implies [40]

$$2\mathcal{T}G(t,s) = \partial_s C(t,s) - \langle \psi_t \mathcal{F}(\psi_s) \rangle \tag{5.16a}$$

$$= (\partial_s - \partial_t) C(t,s) + \langle \mathcal{F}(\psi_t)\psi_s - \psi_t \mathcal{F}(\psi_s) \rangle \,. \tag{5.16b}$$

In some cases (5.16b) can be simplified: for example, if the system is at equilibrium, the last term in (5.16b) vanishes due to the time-reversal symmetry. The other term is also simplified due to the time-translation invariance, so that the equilibrium FDT is obtained. Another simplifying case occurs when $\mathcal{F}(\psi)$ is linear in ψ; the last term vanishes in this case as well, even if the system is out of equilibrium. The linear FRR (5.11) corresponds to this case.

Here we focus on the case of a non-equilibrium system with nonlinear $\mathcal{F}(\psi)$, governed by (3.25) with the initial condition $S_{\text{init}} = 0$. Consideration of higher-order correlations is required in such a case; in order to obtain a useful relation, we need to express the second term on the right side of (5.16a) somehow with C and G.

With (5.13) regarded as a schematic representation of (4.20) in section IV, we recall that the triple correlation can be expressed with memory integrals in (4.21) by the approximation procedure of DIA and MCT. Using this closure scheme, we evaluate the triple correlation corresponding to $\langle \psi_t \mathcal{F}(\psi_s) \rangle$ in (5.14) or (5.16a), and thereby define

$$H(k,t,s) \stackrel{\text{def}}{=} (\partial_s + D_*^{\text{c}} k^2) C(k,t,s) - \int_0^s \mathrm{d}u\, \tilde{M}^G(k,s,u) C(k,t,u) - \int_0^t \mathrm{d}u\, M^C(k,s,u) G(k,t,u). \tag{5.17}$$

This H corresponds to the right side of (5.16a) and should be a constant multiple of G, unless inconsistency is introduced by the closure scheme. Only after checking this consistency, we

can accept the nonequilibrium FRR based on (5.17) as a potentially useful modification to the equilibrium FDT in (4.16).

In the present case, the closure scheme is found to be consistent in the sense that H, defined in (5.17), satisfies the same equation as G, namely (4.23). To demonstrate it, we operate H with $\partial_t + D_*^c k^2$. In an abbreviated notation, in which the k-dependence is omitted and ν is used as a shorthand for $D_*^c k^2$, we have

$$(\partial_t + \nu)H(t,s) = (\partial_t + \nu)(\partial_s + \nu)C(t,s) - \int_0^s \mathrm{d}u\, \tilde{M}^G(s,u)(\partial_t + \nu)C(t,u) - \int_0^t \mathrm{d}u\, M^C(s,u)(\partial_t + \nu)G(t,u) - M^C(t,s). \quad (5.18)$$

The first term on the right side can be evaluated by operating (4.21) with $\partial_s + \nu$. The second and third terms are treated with a technique involving interchange of the order of integration and renaming of the variables, as is exemplified below in the case of the third term:

$$\begin{aligned}\int_0^t \mathrm{d}u\, M^C(s,u)(\partial_t + \nu)G(t,u) &= \int_0^t \mathrm{d}u\, M^C(s,u) \int_u^t \mathrm{d}v\, \tilde{M}^G(t,v)G(v,u) \\ &= \int_0^t \mathrm{d}v \int_0^v \mathrm{d}u\, \tilde{M}^G(t,v)M^C(s,u)G(v,u) \\ &= \int_0^t \mathrm{d}u\, \tilde{M}^G(t,u) \int_0^u \mathrm{d}v\, M^C(s,v)G(u,v). \end{aligned} \quad (5.19)$$

Evaluating all the terms on the right side of (5.18) in this way, we find that many terms cancel each other, leaving a rather simple expression:

$$[\text{right side of (5.18)}] = \int_s^t \mathrm{d}u\, \tilde{M}^G(t,u)H(u,s).$$

Thus H satisfies, in the full notation,

$$(\partial_t + D_*^c k^2)H(k,t,s) = \int_s^t \mathrm{d}u\, \tilde{M}^G(k,t,u)H(k,u,s), \quad (5.20)$$

which duplicates (4.23) with G replaced by H. This means that H is a constant multiple of G. The constant of proportionality, corresponding to $2\mathcal{T}$ in (5.16a), is given by the last term on the right side of (4.22), so that we have

$$\frac{2D_* k^2}{L^2} G(k,t,s) = H(k,t,s) \quad (5.21)$$

with H defined by (5.17). Although this relation is very complicated, it certainly allows determination of G when C is given, at least in principle.

Finally, considering a linear combination of (4.21) and (5.17) corresponding to the tran-

sition from (5.16a) to (5.16b), we obtain

$$\begin{aligned}\frac{2D_*k^2}{L^2}G(k,t,s) &= H(k,t,s)\\ &= (\partial_s-\partial_t)C(k,t,s)\\ &\quad+\int_0^t \mathrm{d}u\,\left[\tilde{M}^G(k,t,u)C(k,u,s)-M^C(k,s,u)G(k,t,u)\right]\\ &\quad+\int_0^s \mathrm{d}u\,\left[M^C(k,t,u)G(k,s,u)-\tilde{M}^G(k,s,u)C(k,t,u)\right]. \end{aligned}\tag{5.22}$$

In this non-equilibrium FRR, the memory integrals represent triple correlations due to the nonlinear term in (3.25). If this nonlinear effect is negligible, (5.22) is reduced to (5.11). In other words, (5.22) could be used to establish the validity range of (5.11) by checking on what conditions the integral terms are negligibly small. In particular, in the limit of large s, the memory integral terms in (5.22) cancel each other, so that the equilibrium FDT is recovered.

VI. CONCLUDING REMARKS

We have presented a theoretical framework for calculation of two-tag correlations in SFD, grounded on the label variable representation of density fluctuations, i.e. (3.22) that governs the fluctuating vacancy field $\psi(\xi,t)$. This framework allows us to calculate the two-tag displacement correlation $\chi_{ij}(t,s)$ and the corresponding response function, $g_{ij}(t,s) = \partial_t\chi^+_{ij}(t,s)$. In the equilibrium case, the Einstein relation between χ_{ij} and χ^+_{ij} has been demonstrated, not only through concrete calculations but also through a link to the "model B" equation (4.17) for which the FDT between C and G is generically known to hold at equilibrium. By applying the above-mentioned framework to the ageing SFD studied by Leibovich and Barkai [22], we have extended their observations on violation of the Einstein relation to the two-tag quantities: the Einstein relation between χ_{ij} and χ^+_{ij} is violated because χ_{ij} is sensitive to the initial condition while χ^+_{ij} is not (within linear approximation). By switching to C and G, we have seen a relatively simple non-equilibrium FRR in (5.11) which is valid in the linear case. Nonlinear analysis by means of the Lagrangian MCT is also discussed.

The everlasting effect of the initial condition on SFD [22] is understood by considering that χ_{ij} given by the Alexander–Pincus formula (3.39) is dominated by the long-wave modes. Since $C^0(k,s)$ has the relaxation time $(D^{\mathrm{c}}_*k^2)^{-1}$ that diverges for $k\to+0$, it takes infinitely long time to equilibrate the single-file system through the time evolution. This observation contains a practically important matter that requires attention when one performs numerical simulations of SFD: If simulation of a system at equilibrium is intended, and if the system is prepared by an equilibration run with a certain finite length of waiting time s, the temporal span of the collected data, $\max t_\Delta$, should never exceed s [38]. A result for a longer span will expose insufficiency of equilibration.

Several possible future directions may be mentioned. One may improve the linear analysis on the ageing SFD in subsection V.B by evaluating the memory integrals in the Lagrangian MCT. The nonlinear FRR in subsection V.C will prove useful as a part of such calculations. In particular, it would be interesting to clarify whether G is completely independent of the ageing effect, with the memory integrals in (5.22) taken into account. Even approximate

evaluation of the integral terms will allow us to establish the validity range of the linearized nonequilibrium FRR (5.11); this kind of nonlinear analysis may be also applicable to other forms of correlations, such as (111) in [36] which reads $(\partial_s - \partial_t)\langle R_i(t,0)R_i(s,0)\rangle$ in our notation. Such calculations will provide methodological insight into the Lagrangian MCT in higher dimensions [54].

As another interesting direction, one may consider SFD-like dynamics with overtaking, which has been studied both in continuous spaces [82–85] and on lattice geometries [86, 87]. In the case of (non-ideal) SFD in the 1D continuous space, finite $V_{\max}$ allows overtaking; the group of the present authors studied the effect of overtaking on the displacement correlation in a previous work [88], and recently the present authors studied the effect on the two-tag elongation correlation [52]. It appears quite promising to study response functions corresponding to these correlations. Finally, it will be interesting to test the validity of the Dean-Kawasaki description of SFD armed with the Lagrangian MCT, which seems approximate but more powerful than the Edwards–Wilkinson theory, by applying it to a number of interesting problems that have studied with other approaches, such as the probability distribution of two-tag displacements [51] and multi-tag correlations [50].

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